Enhancement of Lithium Niobate nanophotonic structures via spin-coating technique for optical waveguides application

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Abstract. This work is dedicated to investigation of temperature effects in Lithium Niobate (LiNbO3) nanostructures. The LiNbO3 nanostructures were deposited on glass substrate by spin-coating technique. LiNbO3 was set down at 3000 rpm for 30 sec and annealed from 100 to 600 °C. The structures were characterized and analyzed by scanning electron microscopy (SEM) and ultraviolet visible (UV-vis) spectrophotometer. The measured results have showed that by increasing annealing temperatures, the structures start to be more crystallized and be more homogenized until the optimum arrangement was achieved. Once this was accomplished, it’s applicable for optical waveguides development. Eventually, it starts to be less crystallization and non-homogeneous. Energy gap was recorded to be at average value of 3.9 eV.

1 Introduction

Lithium Niobate (LN) is a very important optical material which is widely used by the photonics industry mainly due to its excellent electro/acousto-optical properties [1-3]. Lithium niobate (LiNbO3) is an important ferroelectric material because of its excellent piezoelectrical, electrooptical, pyroelectrical and photo-refractive properties [4, 5]. It’s widely used as a polar material for photonic applications [6, 7]. In addition it’s employed in nonlinear optics for frequency conversion and in telecommunication for electro-optic modulation [8, 9]. Furthermore, it is an attractive material for the fabrication of optical wave-guide devices [10, 11]. Moreover, direct and indirect energy band gap of LiNbO3 is reported to be in the range of 3.5 - 4.7 eV which depends on LiNbO3 concentrations. These changes are attributed to several parameters like grain size, composition and defects [12-14]. There are different known approaches to synthesize undoped LiNbO3 nanocrystals using soft-chemistry [15], pulsed laser deposition [16, 17], RF sputtering [18] and hydrothermal methods [19, 20]. LiNbO3 is also studied in its heterojunction structure, photonic crystal and single phase thin film [21-25], and surface acoustic wave device (SAW) [26]. LiNbO3 waveguides are employed broadly in many functional electro-optic and acousto-optic devices [27]. The waveguide structures are essential in many integrated-optic devices especially at high frequency region. This work reports the preparation of LiNbO3 nanostructures by exploiting the spin-coating technique. The characterization and analysis have been elaborated as a function of LN’s annealing temperatures. The refractive index analysis is the main work on LiNbO3 optical waveguides because of refraction coefficients between the base and deposit samples will ensure access to total internal reflection that gives better realization of optical waveguides. The refractive index is measured and calculated in order to fit the best application of optical waveguides. Thus, the focus in the study is to prepare the LN thin film and to enhance towards a high quality film with optical properties closely like a single crystal LiNbO3 wafers.

2 Experimental details

LiNbO3 films were prepared mainly by adding Nb2O5 (ultra-pure, 99.99%) and citric acid (CA) without further purification. The solution was then mixed with Li2CO3...
and Ethylene Glycol. The molar ratio between Li2Co3 and Nb2O5 was 1:1 in order to maximize the formation of LiNbO3 stoichiometry phase. Firstly, the Li2Co3, Nb2O5, and citric acid were dissolved in Ethylene Glycol with simultaneous heating and stirring at 90 °C for 48 hours. Obtaining homogeneous and crack-free films of LiNbO3 requires the precursor to be deposited by spin coating technique on quartz substrates at a spinning speed of 3000 RPM for 30 Sec and 0.5 M/L molarity concentration. In total, seven layers were prepared. The film was further dried at 120 °C for 5 min and calcined at 250 °C for 30 min in static air and oxygen atmosphere with the intention of removal the organics. Finally, the film was annealed from a constant step variation of 100 to 600 °C. The structural evolution of the as-prepared thin films was examined using a high-resolution X-ray diffraction (HR-XRD) system equipped with Cu-Kα radiation of wavelength λ = 0.15418 nm at 40 kV and 30 mA. The thickness of the annealed samples was measured using scanning optical reflectometry. The scanning electron microscopy (SEM) was employed to investigate the surface morphology of LiNbO3 and the atomic force microscopy (AFM) was utilized to observe the surface roughness of the LiNbO3 films. The optical properties were inspected by means of double-beam Ultra-Violet (UV-vis) spectrophotometer.

3 Results and discussion

3.1 Morphological studies

Figure 1 shows SEM images of LiNbO3 nanostructures deposited on quartz substrates at different annealed temperatures. The density of nucleation for the LiNbO3 thin films was not uniform on the flat substrate with dissimilar temperatures. At least molarity concentration notifies the emergence of a high proportion of the pores and voids. These pores were observed as a result of impurities impacts like Nb2O5. The LiNbO3 thin films grew smoothly at 500 °C that’s led to good distribution better than other annealing temperatures as shown in Fig. 1. On the other hand, the structure is more homogenous at higher molar concentration [14]. This suggests that 500 °C leads to increase the regular distribution of LiNbO3 nano and micro structures. A closer examination of these samples indicates that our synthesized film like ice layers shape of morphology.

![Fig. 1. Surface morphology of LiNbO3 nanostructures with different annealing temperatures. (a) 400 °C, (b) 500 °C, and (c) 600 °C](image)

3.2 Optical properties

The transmission spectra of LiNbO3 nanostructures at different annealing temperatures are shown in Fig. 2. It was measured that the transmission increased with the annealing temperature. This may be due to the increasing of grain size and decreasing thickness. Similar result was also recorded in other work [28]. The values of transmission were about 70-80% with the annealing temperature range of 100 to 600 °C. The deposited samples were white to brown in color and showed high transmittance. The high value of transmittance is attributed to the excessive (LiNbO3) ions at interstitial site that increased its transparency level.
Figure 3 shows the reflectance of LiNbO₃ nanostructures in the wavelength range of 250-800 nm at room temperature. The reflection spectra were found to decrease with annealing temperature due to the increment in the surface roughness that related to crystals grain size.

The energy band gap ($E_g$) was established as a function of photon energy in Fig. 4. All curves were singularized at a specific point around 3.25 eV. However, the $E_g$ was extrapolated and recorded at different values. The value was noted to increase with annealing temperatures due to shrinkage in the grain size.

The refractive index ($n$) was determined from a transmittance spectrum as a function of the wavelength at different annealing temperatures. The spectrum showed that there was a decrease in the refractive index with incident wavelength from 2.32 to 2.56 which related to the drop in the reflectance as depicted in Fig. 5.

Beside that an increment in the refractive index could be realized in the UV region with the annealing temperature as shown in Fig. 6. This behavior may be incorporated to growth in packing density as a result of the oxidation process during heat treatment. All the higher values of refractive index are suitable for optical waveguide fabrication.

4 Conclusion

The LiNbO₃ nanostructures have been chemically prepared by spin-coating technique. SEM shows diameter of grain size from 64 to 167 nm. Optical properties give high values of transmission which is about 89 - 96% and the measured energy band gaps are 3.6, 3.85, 4 and 4.2 eV. We found an approximate match between the energy band gap in the values calculated using the transmission spectra. The refractive index is determined from the transmission spectrum and established its appropriateness for application in optical waveguides that needs high value.

References